In-lake neutralisation and post-rehabilitation treatment of a lignite mining pit lake – lessons learned

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Abstract The formerly highly acidic pit Lake Bockwitz south of Leipzig (Saxony) has been repeatedly treated since 2004 with soda ash to meet the regulatory water quality standards for the lake effluent. Intense monitoring of certain water quality parameters and budget calculations showed that the loads of acidity into the lake were underestimated. To identify the responsible processes of re-acidification, field research was extended to quantify the acid flux via groundwater inflow, sediment interaction, groundwater recharge, post-acidification of bank substrate and its erosion into the lake. Field monitoring combined with bench experiments enabled us to determine the key input variables for the adequate application of geochemical and hydrogeological models. The results indicate that groundwater recharge coupled with post-acidification of the bank substrate was the main factor of re-acidification. These investigations are proved to be critical to reliably predict the future water quality of mining pit lakes, to develop optimal treatment measures for the post-rehabilitation period, and to plan the financial needs for cost-effective lake water conditioning.

Key Words pit lake, acid load, in-lake neutralisation, monitoring, groundwater recharge, geochemical modelling

Introduction

The reclamation of former open-cast mining areas is usually coupled with flooding of the abandoned open pits to establish a self-regulating water balance and develop lakes for recreation, nature conservation, and water management. Depending on the intended use of the lake and its discharge into a connected river basin, the objectives of the European Water Framework Directive for water quality parameters have to be met. A key objective is to prevent deterioration of the fresh water quality. In particular, discharge of acidity (monitored by the pH value), salt load, and heavy metals including iron (Fe) are limited. However, most lignite mining pit lakes do not meet the anticipated water quality due to pyrite weathering in the overburden substrate and acidity generation in connected aquifers (Schultze *et al.* 2010). Hence, water treatment measures are applied ex situ (e.g., treatment plants for discharge) or in situ (e.g., in-lake neutralisation). This paper presents results and conclusions from 7 years of in-lake neutralisation and post-rehabilitation treatment of Lake Bockwitz using soda ash (Na_2CO_3).

The former open-cast lignite mine Borna-East, situated south of Leipzig (Germany), was operated from 1850 through 1990. After closing the mine and stopping the dewatering, Lake Bockwitz filled by natural groundwater rise between 1993 and 2005 as the largest lake in a series of lakes that formed within the abandoned mining area. Lake Bockwitz has a maximum depth of 19.5 m, a volume of about $18.7 \cdot 10^6$ m³ and a surface area of ≈ 1.7 km²; its water table is at ≈ 146 m a.s.l. To keep the water table constant, surplus water is drained off into the Eula and Wyhra river since 2006.

The drain water is monitored for compliance with quality criteria (pH >6, total Al <0.5 mg/L, total Fe <3 mg/L). Therefore, the mining administration authority LMBV has accomplished a pilot project from 2004 to 2006 to neutralise the strongly acidified lake (pH 2.7, Al_{tot} ≈ 19 mg/L, Fe_{tot} ≈ 55 mg/L) using soda ash. The initial supply of Na_2CO_3 was calculated from a limnological forecast on the lake's quality by Guderitz *et al.* (2003). Linked to this work, a standardised data collection, analysis and quality control directive for mining-related hydrological monitoring (MHM) was developed (LMBV 2007). However, since the annual acidity load to the lake has initially be considered minor (less than 10% of the entire alkalinity supply), the present study aims at quantifying the specific sources of acid inflow to the lake to improve the operational treatments during the post-rehabilitation period. Hydrogeological and geochemical models are applied and optimised as forecast tools. These models are based on parameters continuously gathered by the process-oriented MHM monitoring network.

Material & Methods
Field Methods
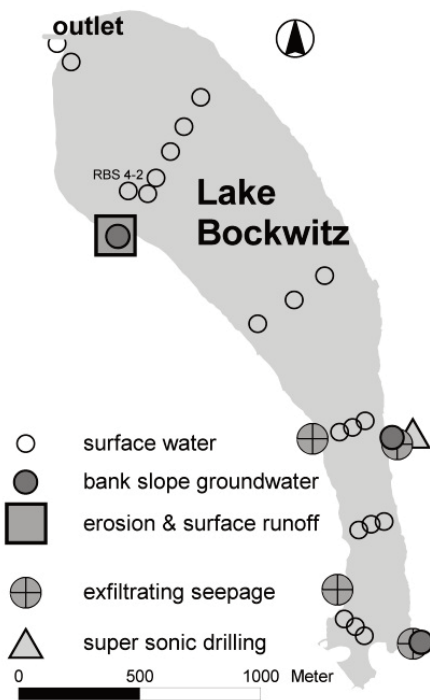All sampling and field operations as well as analytical methods followed the MHM code of practice

Figure 1 Sketch of Lake Bockwitz monitoring site, locations of sensing and sampling sites.

(LMBV 2007). This standard guarantees high quality data and their comparability among other monitoring networks within the LMBV scope of responsibility.

Lake water and sediment composition were monitored quarterly near the maximum depth. Additional water and sediment samples were taken fortnightly or monthly at distinct locations and water depths along the longitudinal axis. Water composition and discharge of the main inlet and outlet of Lake Bockwitz were monitored on a monthly basis.

Data on long-term groundwater composition of specific aquifers were obtained from the groundwater monitoring network operated by the LMBV since 1993. These permanent wells are sampled and analyzed once a year according to the MHM standard. The same sampling strategy was adapted to six temporary monitoring wells set up close to the shoreline. These wells enabled us to determine the actual groundwater flow to the lake and study the recharge-driven change of groundwater composition on its transit through the overburden substrate. To collect seepage water (from interflow and groundwater recharge) out of the bank slopes, three water samplers have been installed and sampled six times in 2010. The set of analytical parameters was analogous to the monitoring wells.

Acid and alkaline input through surface runoff and erosion were quantified on a 30 m² field plot by an automated erosion recording and sampling system. Depending on the recorded quantity of precipitation, the mass/ volume and composition of runoff and eroded matter as well as the soil moisture content were determined. Supersonic drilling was applied to explore the stratigraphic structure of the bank slopes from the surface to the aquitard. At different depths soil columns were isolated for laboratory tests and soil eluates prepared to measure pH value and electrical conductivity on-site.

Laboratory Methods
Water and soil samples were characterized with respect to a variety of physical and chemical parameters according to the MHM standard procedures (LMBV 2007). The acid release capacity of the bank substrates was tested after oxidation with hot 30% H_2O_2 solution in batch slurries. Two soil columns collected from representative slope environments were treated differently to mimic the acid leach-

Table 1 Investigated system components, sensing devices and number of locations for measuring acid input fluxes.

System Component	Sensing Device	Scope of Investigation	Number
Surface runoff	automatic event-driven runoff sampler	water quantity, quality	1
Erosion	automatic event-driven runoff sampler	solid matter quantity, quality	1
Interflow	monitoring wells	water quality, level	6
Groundwater	monitoring wells	water quality, level, vertical plots	9
	super sonic drilling	stratigraphic soil sampling 30m depth	1
	soil column tests	soil physics, water quality, time dependence	2
Lake water	sampling locations	water quality by depth, vertical plots	17
Sediment	sampling locations	composition by depth, soil physics	17

ing of uncovered overburden material due to groundwater flow or infiltrated rain water. While column from the vadose zone was infiltrated with aerobic synthetic rain water, anaerobic groundwater from the field site was used for the column obtained from the saturated zone. Up to ten pore volumes were exchanged by intermittent infiltration flow; i.e., the pump speed was adjusted to the exchange of one pore volume within the first 24 h, followed by 24 h of stagnancy. This procedure aimed at simulating the release of acidity from the bank substrates due to pyrite oxidation and subsequent elution of acid reaction products.

Modelling

Lake Bockwitz is included as a mixed compartment in a cascade of basins for which a hydrochemical mass balance model has been developed (Guderitz *et al.* 2003). Water balance and mass flow of all components including influents and effluents have been validated with respect to hydrology and conservation of mass.

Monitoring of erosion provided a set of parameters for the modelling code E3D (Schmidt 1996). This model quantifies rainfall induced soil erosion in catchments based on physically founded, process oriented algorithms. The model was calibrated and validated for the monitoring plot using rates of surface runoff and loss of matter recorded during the 2008/2009 sampling period. A GIS based relief analysis and detailed hydrotope mapping enabled an upscaling of the processes for the whole catchment of the lake by calculating the distribution and concentration of the runoff including detachment, transport and deposition of particles, in particular for the bank slope along the shoreline.

Using water quality and level data of essential groundwater inflows derived from the regional

groundwater simulation on PCGEOFIM (Müller *et al.* 2003) and the results of the laboratory experiments, a water flow model based on the software HYDRUS3D (Šimůnek *et al.* 2006) was adapted to Lake Bockwitz to calculate other sources of acidity like groundwater recharge, infiltrating, and exfiltrating seepage of the bank slopes. A transient 2D hydraulic transport model for aquifer conditions was set up with geometry of the bank slopes, surface discharge coefficients, soil-hydraulic parameters of stratigraphic material distribution. This model was used to determine groundwater recharge, interflow and exfiltration at lower parts of bank slopes combined with time-dependent generation of acidity. The hydraulic transport model was parameterised for partially saturated bank slopes using experimentally determined soil moisture tension and saturation curves.

Results and Discussion
In-Lake Neutralisation

Initial neutralisation of Lake Bockwitz by powdered soda ash (Na_2CO_3) started in March 2004. The material was blown into the lake just below its surface through a floating pipeline located in the southern part of the lake (Rönicke *et al.* 2010). The highest soda amounts had been applied within the first two years (Figure 2). Since then, operational soda supply was calculated from the ongoing consumption of the lake water alkalinity. In 2007, the floating pipeline was moved from the south to the north side close to the outlet of Lake Bockwitz. Up to the end of 2010 more than $17 \cdot 10^3$ metric tons of soda have been added, thereof 80% from the south and 20% from the north side of the lake. Between 2004 and 2006, about 65% of the added soda was consumed for the neutralisation of the lake water, raising pH value up to ≈ 5. The pH decrease in 2006 showed that roughly 10%
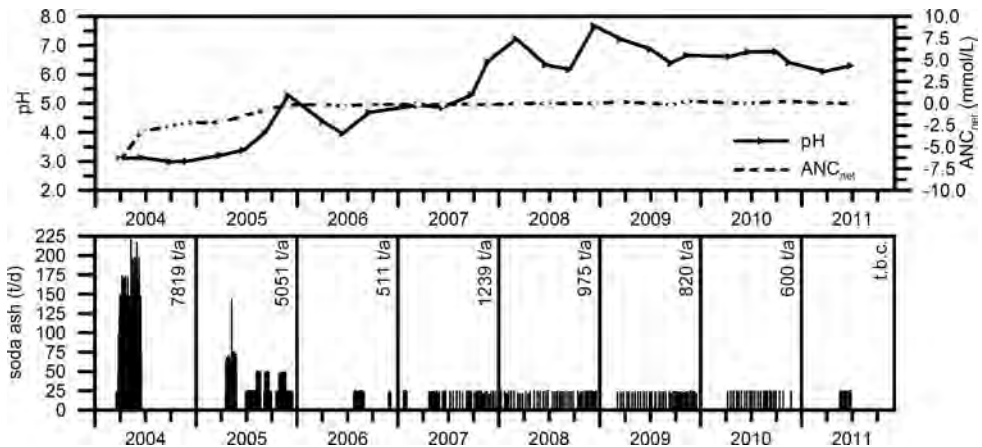

Figure 2 *Net alkalinity (ANC$_{net}$) and pH value as a function of time (sampling station RBS 4—2) in response to annual soda supply shown in metric tons per day (bottom).*

of the added alkalinity got lost by acid inflow from other pit lakes located upstream and from adjacent overburden dumps (Neumann *et al.* 2007). In November 2007, a low positive net alkalinity was reached for the first time. To maintain a moderate level of net alkalinity (ANC_{net}) while reducing the treatment costs, soda applications were sustained by continuously lowering the soda quantity up to date.

Post-Rehabilitation Studies

Mass transport into the lake by erosion was found to take place on sparsely and non-vegetated areas that comprise about 11% (0.64 km²) of the total catchment. The total surface runoff yielded $29 \cdot 10^3$ m³ in 2007 and $380 \cdot 10^3$ m³ in 2008, delivering an annual net acidity (ACY_{net}) of 15 kmol (2007) and 200 kmol (2008). Consistent with this high variability, input of solid matter and associated net acidity amounted to 3.0 metric tons with 0.6 kmol in 2007 and 94.0 metric tons with 20.0 kmol in 2008, respectively.

Field data of groundwater quality close to the surface and close to the shoreline (from temporary wells on the bank slopes) indicate high loads of acidity from groundwater, affected by pyrite weathering and interflow processes. Although the values of net acidity were highly variable within the monitoring period from 2007 to 2009, a clear trend showed that close to the shoreline, the acid load was up to 20 times higher than from upstream aquifers. The highest groundwater flux (≈ 61% of all groundwater inflow) came from the southeastern tertiary aquifers (Guderitz *et al.* 2003). Between 2007 and 2009, ACY_{net} mean values were ≈ 6.0 mmol/L for the vadose zone and ≈ 1.6 mmol/L for the saturated zone. The eastern aquifers yielded lower fluxes, but higher concentrations (vadose zone: ≈ 19.9 mmol/L, saturated zone: ≈ 6.2 mmol/L). Seepage from the western shoreline appeared less significant (≈ 6% of all groundwater inflow; Guderitz *et al.* 2003). The ACY_{net} values of this aquifer showed a decreasing trend; on average we measured ≈ 10.3 mmol/L for the vadose zone and ≈ 0.2 mmol/L for the saturated zone.

Supersonic drilling and on-site analyses of soil eluates combined with acid release tests using H_2O_2 solutions indicated a high potential of acid release for each stratigraphic unit. During the experiments, all soil samples collected from 10 m to 21.5 m below the surface showed a decrease of pH from ≈ 7.5 to ≈ 2.5 on average.

Different results were obtained from the intermittent column experiments testing oxic vadose and anaerobic saturated conditions (Figure 3). For the vadose zone column, the release of net acidity ($ACY_{net\ vad}$) decreased from 12.6 to 1.5 mmol/L after the second pore volume exchange. Continued ex-

changes of the pore volume yielded a steady $ACY_{net\ vad}$ of ≈ 1.3 mmol/L. The pH_{vad} rose from initially 2.9 to a mean value of ≈ 5.6 maintained for exchanged volumes 3 to 10. For the saturated zone column, an almost constant release of net alkalinity ($ACY_{net\ sat}$ = -2.2 mmol/L) and a mean pH_{sat} of 7.6 were found (Figure 3).

The acid inflow from vadose and saturated bank slopes is driven by the transient physical and chemical interactions of groundwater recharge, interflow, exfiltration, and groundwater flow. For a precise forecast of the input of acid into Lake Bockwitz, the results of groundwater monitoring, soil column tests, and calculated vadose and saturated water fluxes need to be combined by a transient model of surface and subsurface flow processes on the bank slopes. The interaction of all significant processes for which fluxes have been calculated is summarized in Figure 4.

Using a physically founded transient 2D flow model, the precipitation infiltrating the soil was discriminated with respect to interflow and groundwater recharge. Of the infiltrating oxic water, 90% acted as interflow stimulating the pyrite weathering process within the vadose zone layers. The remaining 10% of the infiltrating water generated groundwater recharge at the bank slopes. The concentrations of net acidity measured at groundwater wells of the vadose interflow zone were assigned to the transport calculations. The average loads of net acidity to the lake were $ACY_{net\ vad}$ ≈ 24 kmoL/d by interflow and ≈ 2.6 kmoL/d by groundwater recharge. The latter acid influx was completely compensated by the high flux of net alkalinity up to ANC_{net} ≈
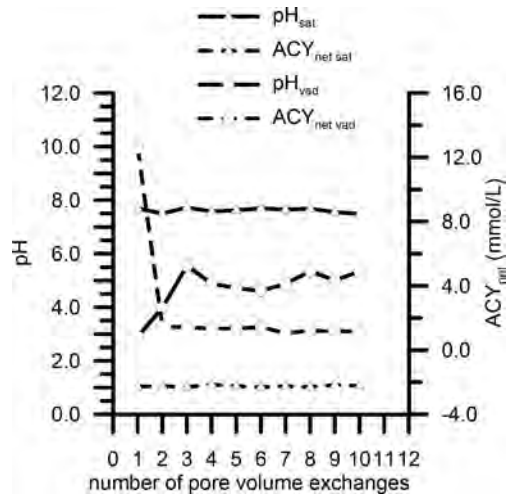

Figure 3 pH and net acidity of the effluent from intermittent soil column experiments performed on an oxic vadose zone sample (vad) and an anaerobic saturated zone sample (sat).
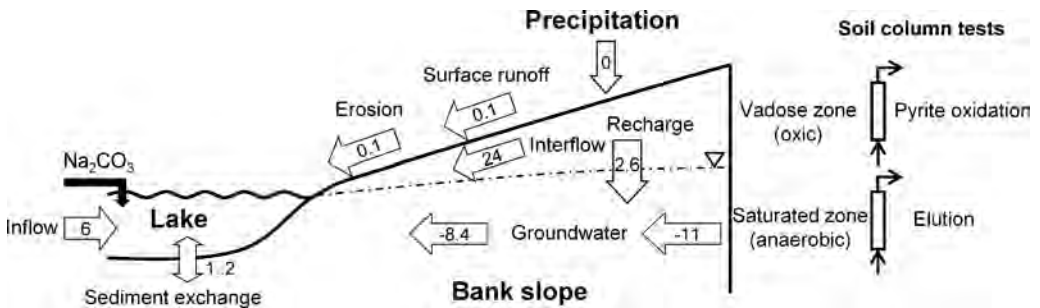
Figure 4 Investigated system components and calculated flows of acidity (positive) and alkalinity (negative numbers, in kmoL/d) to Lake Bockwitz after its neutralisation. Acid release in the vadose and saturated zones was determined by soil column tests.

8.4 kmoL/d entering the lake through groundwater. Surface water discharge from small ponds and dumps upstream of Lake Bockwitz delivered an acid flux of $ACY_{net} \approx 6.0$ kmoL/d by an inlet stream. The acid release from sediment layers at the bottom of the lake is not finally validated. The expected range is $ACY_{net} \approx 1.0$ to 2.0 kmoL/d (Figure 4).

Conclusions

After soda ash has been successfully applied for in-lake neutralisation of Lake Bockwitz in 2004 and 2005, re-acidification proceeded faster than initially predicted from groundwater flow studies. By combination of standardised, process oriented field monitoring with lab experiments, reaction and transport modelling, several sources of acidity and alkalinity have been quantified. Due to continued pyrite oxidation within the overburden substrate, interflow to the lake has been identified as the primary source of acidity. Acid transport by recharge of groundwater is more than balanced by the alkaline groundwater flow to the lake. Runoff from the bank slopes and erosion of acid reacting solids are negligible. Based on the monitoring results from the investigated area since 2007, the demand of soda was predicted more precisely and reduced between 2007 and 2010. Modelling tools proposed in this work allow a more sophisticated prediction of future acid-base budgets. Further post-rehabilitation measures like barrier layers on top of bank slopes of open pit areas to decrease interflow quantity can be designed.

Acknowledgements

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